Highly hierarchical porous structure constructed by NiO nanosheets act as Li-ion and O$_2$ path way for long-cycling rechargeable Li-O$_2$ batteries

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1. Experimental Section

Preparation of NiO

Flower-like NiO were synthesized as follows: 1.502 g CO(NH$_2$)$_2$, 1.314 g NiSO$_4$·6H$_2$O and 0.370 g NH$_4$F were dissolved in 50ml deionized water under stirring at room temperature for 5 h. Then the mixture was transferred into a Teflon-lined autoclave and kept at 120 °C for 2–5 h, and natural cooling to room temperature followed. After washed by deionized water and ethanol for several times, the chartreuse products (Ni(OH)$_2$·0.75H$_2$O) were collected, and then dried at 80 °C in an oven for 24 h. At last, the gray products (NiO) were obtained by calcinating the light green precursors at 600 °C for 2 h in the air.

Characterization

The crystal structure of the samples was checked by X-ray diffraction (XRD, Bruker D8 with Cu K$_\alpha$ Radiation 40kV, 40mA) and the morphology of the products were characterized by scanning electron microscopy (SEM, SU-70 and LEO-1530) and transmission electron microscopy (TEM, JEM 2100, 200kV). The thermogravimetric analysis (TGA) measurement was performed by SDT-Q600 thermal analyzer with 10 °C·min$^{-1}$. 
Electrochemical analysis

2032 type coin cells were assembled with lithium metal as the counter electrode. The slurry was prepared by mixing the flower-like NiO, Ketjen carbon and polyvinylidene difluoride (PVDF) in a weight ratio of 80:10:10. The slurry was pasted on a carbon paper and dried at 80 °C for 12 h in a vacuum oven. The mass loading of NiO/KB/PVDF was about 1.0-1.3 mg·cm\(^{-2}\). The electrolyte was 1M LITFSI (lithium bis-(trifluoromethanesulfonyl)-imide) in TEGDME (tetraethylene glycol dimethyl ether). The amount of electrolytes for each cell was 100 uL. The battery with glass filter paper as separator was conducted in an Ar filled glove box (<0.1 ppm of H\(_2\)O and 0.1 ppm of O\(_2\)). The Li-O\(_2\) battery was stored in a box filled with pure O\(_2\) (99.999%) when tested. Electrochemical impedance spectroscopy investigations were checked on an Autolab 1.9 electrochemistry workstation. The NEWARE battery test system was used to test the galvanostatic discharge/charge capacities at different current densities in the potential range of 2.0 V to 5.0 V. The galvanostatic discharge/charge test was based on the mass of KB.

2. Figures

![Figure S1. The XRD pattern of Ni(OH)\(_2\)·0.75H\(_2\)O (up) and NiO (down)](image-url)
Figure S2. The TGA curve of Ni(OH)$_2$$\cdot$0.75H$_2$O from room temperature to 600 ºC in air atmosphere.

Figure S3. The nitrogen adsorption–desorption and pore size distribution (inset) curves of (a) precursor (Ni(OH)$_2$$\cdot$0.75H$_2$O) and (b) NiO.
Figure S4 Discharge/charge curve of Li–O$_2$ batteries made by KB cathodes at a current density of 200 mA·g$^{-1}$ for the different cycles (a); Discharge/charge curves of Li–O$_2$ batteries made by NiO cathodes at a current density of 200 mA·g$^{-1}$ for the different cycles (b) and Discharge/charge curve of Li–O$_2$ batteries made by KB and NiO cathodes at a current density of 200 mA·g$^{-1}$ for the first cycle; the cycle number of KB and NiO cathodes at 200 mA·g$^{-1}$ with a limited capacity of 1000 mAh·g$^{-1}$ (c).

Figure S5. The rate performance curves of Li–O$_2$ batteries made by NiO cathodes
with a current density of 100 mA·g⁻¹, 200 mA·g⁻¹, 500 mA·g⁻¹ for the first cycle, respectively.

![Figure S6. The EIS patterns of Li-O₂ batteries with different cycles at the current density of 500 mA·g⁻¹ indicated that the charge transfer impedance increased with the cycles number.](image)

<table>
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<th>Materials</th>
<th>Capacity (mAh g⁻¹)</th>
<th>Current density</th>
<th>Cycle number</th>
<th>Ref.</th>
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<tr>
<td></td>
<td>200 (limited)</td>
<td>100 mA g⁻¹</td>
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<td>our experiments</td>
<td>1000 (limited)</td>
<td>200 mA g⁻¹</td>
<td>80</td>
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Table S1 The electrochemical performance of NiO by the other reports

Reference
